

# Thermal-spikes temperature measurement in pure metals under argon ion irradiation ( $E = 5\text{-}15\text{ keV}$ )

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**Abstract.** A method for determining the parameters of energy release in dense cascades of atomic displacements and for estimating the temperature and the level of pressures in thermalized cascade regions (thermal spikes) is proposed. It is based on the measurement and analysis of the spectral composition of the surface glow of condensed media in the course of irradiation with  $\text{Ar}^+$  5-15 keV accelerated ions, under the assumption of the presence of a thermal component of glow defined by the presence of thermalized cascade regions.

## 1. Introduction

Many authors observed the surface glow of various substances in the visual range under the action of accelerated ion beams. The emission spectra displayed both discrete lines and continuous bands. When irradiating metals, the presence of oxides on their surface should be considered. Fortunately, these oxides are sputtered by continuous ion beams ( $P_s > 1\text{ W/cm}^2$ ) for a few minutes of exposure.

None of the theoretical models proposed so far, which can be conditionally subdivided into thermodynamic, molecular, detachment, and collisional ones [2, 3], is able to explain all the peculiarities of the luminescence provoked by ion irradiation and describe its spectrum. It was found that the bands of continuous emission does not exhibits some features of luminescence.

In connection with this, to explain the presence of the continuous spectrum (continuum), it can be assumed that we are dealing with some equilibrium (in other words, thermal) or at least quasiequilibrium emission. This can be, e.g., the emission from so-called "thermal spikes" that are formed in the immediate vicinity of the surfaces of solids [4, 5] bombarded by ions of low and medium energy (1-100 keV). These spikes appear as a result of the evolution of dense (nonbranched) atomic collision cascades. This hypothesis is illustrated by Figure 1. Figure 1 also shows the possible effects of nanoscale shock wave generation by heated rapidly expanding regions of thermal spikes. This possibility is confirmed by the data below.

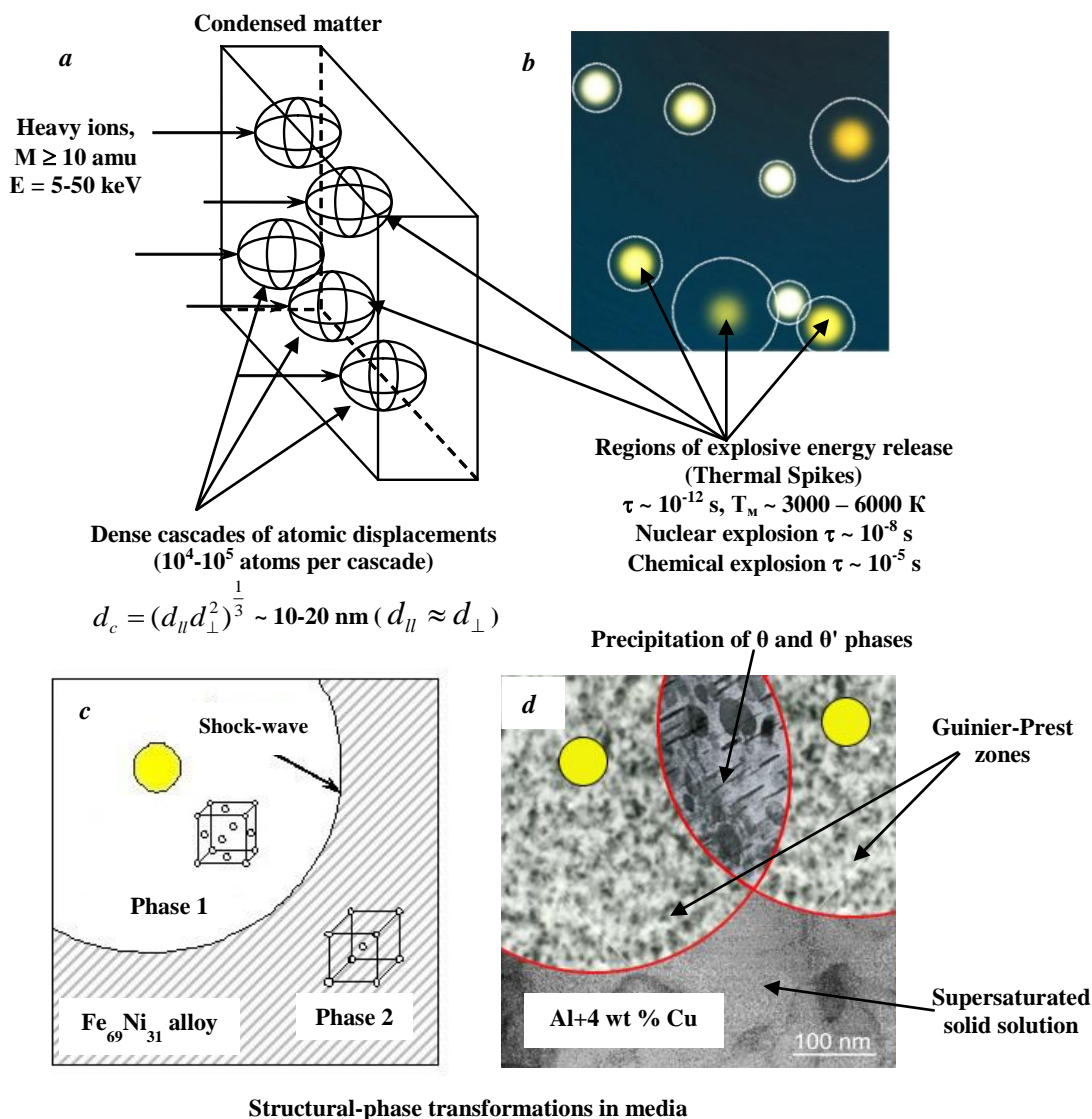
## 2. Experimental

Irradiation was performed with  $\text{Ar}^+$  5-15 keV ions using an ILM-1 ion beam implanter equipped with a PULSAR-1M ion source based on a low-pressure glow discharge with a hollow cold cathode [1].

The emission spectra of the targets in the course of irradiation were measured with a multichannel photodetector based on OC-12 diffraction spectrograph and a CCD array in the range from 360 to 850



nm. We used a quartz optical fiber, the receiving end of which was set at a distance of 1 cm from the edge of the sample and was directed onto the sample surface at an angle of  $60^\circ$ .



**Figure 1.** Radiation-dynamic effects under ion irradiation (c, d illustrate the data of [6] and [7]).

It should be noted that the depth of visible radiation penetration into metals is on the order of  $\lambda/2$  (where  $\lambda$  is the wave length of radiation), which greatly exceeds the depth of the formation and the linear dimensions of thermal spikes.

### 3. Results and discussion

In order to verify the aforementioned hypothesis, we have analyzed the spectral composition of optical emission from high-purity tungsten (99.99 W), iron (99.96 Fe), and commercial aluminum (99.5 Al) bombarded by  $\text{Ar}^+$  ions with energies  $E = 5-15$  keV. It was of interest to compare the experimental temperatures and those calculated using the theory and computer simulations in the field of thermal spikes (as average energy per cascade atom), depending on the energy of the accelerated ions.

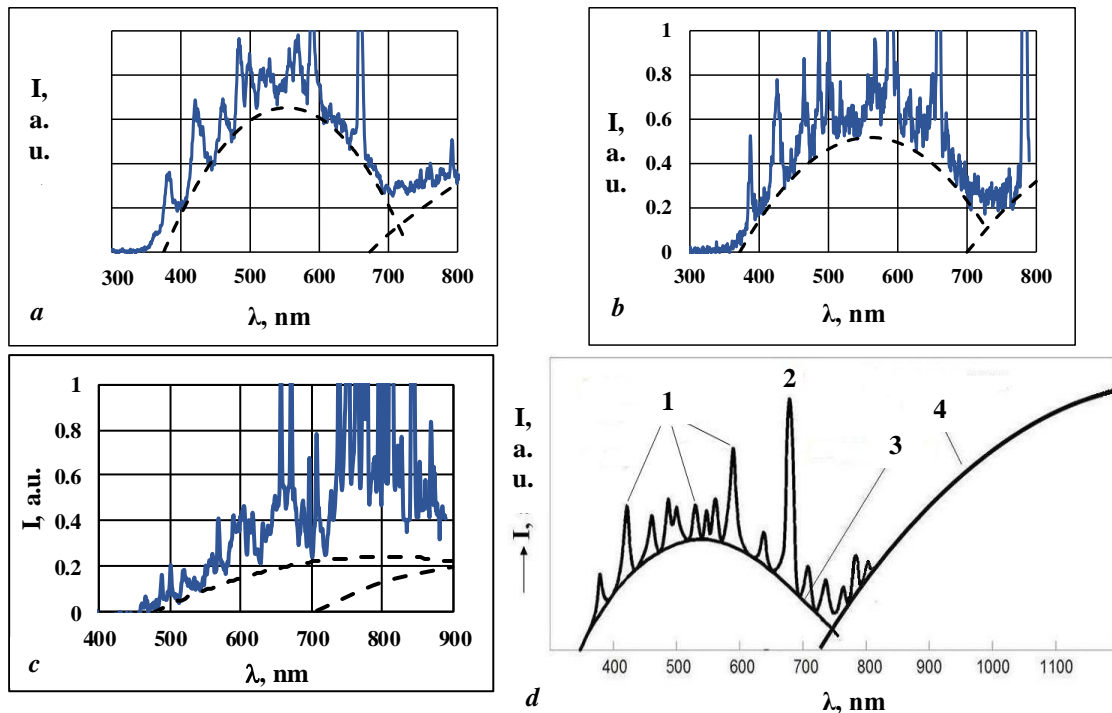
The emission spectra (Figure 2) displayed both discrete lines and continuous bands. The discrete emission can be generated by a fraction of atoms, ions, and molecules dissipated from the surface

layers of the targets being in excited states. Any nonequilibrium emission outside a solid body must be discrete because of the lack of quasi-continuous energy bands. For this reason, none of the proposed so far theoretical models (all of them belong to the different types of luminescence) can explain all the features of this kind of emission, in particular, to describe the continuous component of the spectrum.

In accordance with the above assumption we considered that the spectra of all the targets have two broad bands of Planck thermal radiation (Figure 2), which can be described by the relation:

$$\varepsilon_{\lambda, T_1, T_2} = \varepsilon_0 \cdot \frac{2\pi hc^2}{\lambda^5} \left[ \frac{a_1}{\exp(\frac{hc}{k\lambda T_1}) - 1} + \frac{a_2}{\exp(\frac{hc}{k\lambda T_2}) - 1} \right], \quad (1)$$

where  $h$  is the Planck's constant and;  $c$  is the speed of light in vacuum;  $k$  is the Boltzmann constant;  $T_1 \equiv T$  is the temperature of nano-sized areas of explosive energy release (thermal spikes);  $T_2 \equiv T'$  is the temperature of integrally heated matrix (target) measured by a thermocouple,  $\varepsilon_0$ ,  $a_1$ ,  $a_2$  are the constants taking into account the degree of blackness and integral sections (area) of radiating regions (thermal spikes and matrix) as well as the absorption of the surface layer and instrumental factors.



**Figure 2.** Emission spectra of targets bombarded by  $\text{Ar}^+$  ions: (a)  $\alpha\text{-Fe}$ ,  $E = 15$  keV,  $T = 5270$  K,  $T' = 533$  K; (b)  $\text{W}$ ,  $E = 15$  keV,  $T = 5080$  K,  $T' = 520$  K; (c)  $\text{Al}$ ,  $E = 10$  keV,  $T = 3600$  K,  $T' = 490$  K; (d) schematic diagram illustrating formation of the emission spectra of the irradiating target.

The first band has a maximum at wavelength  $\lambda_{\text{ml}}$ , which varies within 500-805 nm for investigated metals, according to the hypothesis under consideration, corresponds to equilibrium Planck emission from strongly heated near-surface regions (thermal spikes) of ion-bombarded targets. The typical radius of these regions is  $\sim 3\text{-}5$  nm<sup>1</sup>. Perhaps it can also refer to glowing of dense surface plasma

<sup>1</sup> The time of cascade thermalization (i.e., thermal spike formation) is  $\sim 10^{-12}$  s, while the time of its cooling is  $\sim 10^{-11}$  s. There simultaneously exist no more than  $10^4$  of such nano-sized "hot spots" on an area of  $\sim 1$  cm<sup>2</sup> at an ion beam current density within 50-150  $\mu\text{A}/\text{cm}^2$ . The average distance between them is on the order of several hundred microns.

formed during "splashing" of thermal spikes [2, 3], in which the pressure, as shown below can be up to several GPa. This may explain a glowing halo experimentally observed near the target surfaces [2].

The maximum of the second emission band at wavelength  $\lambda_{m2}$  falls in the IR spectral range. This band is only manifested by its short-wavelength wing that is due to thermal radiation from a target integrally heated by ion beam ( $\lambda_{m2}$  was determined based on thermocouple readings).

On the background of these broad bands, all spectra displayed much narrower bands of nonequilibrium emission. Some of these bands corresponded well to the emission from atoms of the bombarded metals and from argon atoms. Figure 2e shows a diagram of the formation of the observed spectral composition of the target emission under bombardment with accelerated ion beams.

In the assumption that the system attains *quasiequilibrium* state in the region of thermal spikes formed as a result of thermalization of the dense cascades of atomic displacements, using Wien's displacement law, we obtain estimations of temperature in the region of thermal spikes as a function of parameters of ion bombardment (Table 1). Estimated errors are about 50-200 K.

A decrease in the temperature of a thermal peak with an increase in ion energy (Table 1) is due to the advanced growth in the cascade volume with respect to the ion energy generating a cascade. Figure 3 shows a comparison of the calculated (using Monte-Carlo TRIM [8] and kinetic Boltzmann equation [9] methods) and experimental values of the temperature of the thermal peaks.

**Table 1.** Results of determination of the temperature and pressure in the region of thermal spikes in metals during their irradiation with accelerated argon ions (E = 5-15 keV)

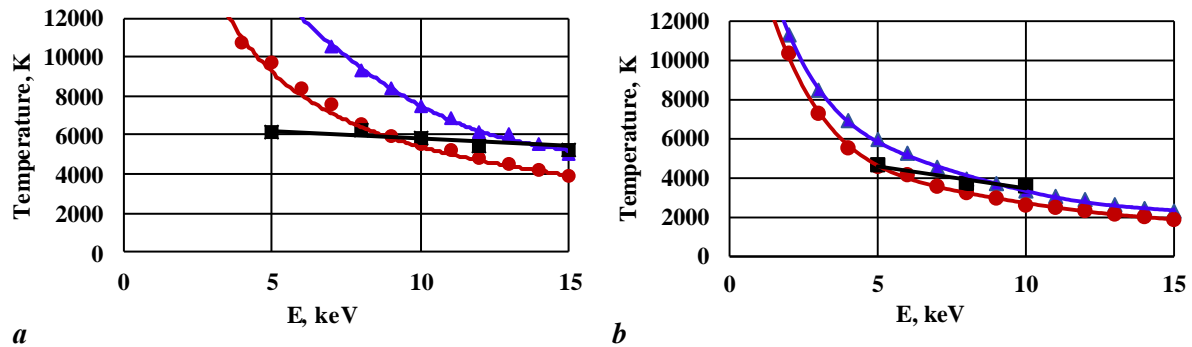
Energy Ar <sup>+</sup> , keV	Wave length of peak position, nm	Target tempera- ture, K	Temperature of the cascade region, K and respective estimated pressure, GPa				Experimental yield strength, GPa	Theoretical yield strength, GPa
E	$\lambda_{m_1}$	$T'$ (exper.)	$T = b/\lambda_{m_1}$ (exper.)	P	T* (TRIM)	P*	$\sigma_T^{real}$	$\sigma_T^{teor} = \left( \frac{\mu}{6} - \frac{\mu}{30} \right)$
Al								
5	620	348	4674	3.67	6007	4.73	0.05-0.120	3.46
10	805	490	3600	2.59	3526	2.54		3.46
Fe								
5	500	343	5800	6.56	13897	15.7	0.1-0.25	9.33
10	535	388	5420	5.97	7569	8.34		9.33
15	550	533	5270	5.50	5250	5.48		9.33
W								
5	505	328	5740	4.71	6566	5.39	0.7-1.4	21.46
10	525	413	5520	4.26	3894	3.01		21.46
15	570	520	5080	3.61	2928	2.08		21.46

As the Table 1 shows, the wavelength of the iron target that corresponds to the maximum of the Planck radiation at an energy of Ar<sup>+</sup> ions of 5 keV is 500 nm. In accordance with Wien's displacement law, this corresponds to T = 5800 K ( $b = 0.002898$  mK is the Wien's constant).

It is easy to calculate that the rate of energy release in dense cascade of atomic displacements with a radius of about 3-5 nm (a form of cascade in the case of heavy ions is an ellipsoid of revolution with similar semi-axes size) during its thermalization ( $\sim 10^{-12}$  s) is nearly the same as in a nuclear explosion (with a nuclear plasma at  $10^8$  K and characteristic time  $\sim 10^{-8}$  s).

For the target made of commercially pure aluminum, the temperature in region of thermalized cascades (Figure 3, Table 1) is lower than that in heavy metals, that is consistent with the results obtained by the numerical methods [8, 9]. A large excess of the calculated data over the experimental ones for the Fe target at low argon ion energy (and, consequently, extremely small sizes of the

cascade) may be due to the decisive role of the heat removal. At high energies, the opposite effect may be associated with cascade fragmentation, which is not taken into account due to averaging in simulation methods.



**Figure 3.** Theoretical ( $\blacktriangle$ ) [8], ( $\bullet$ ) [9] and experimental ( $\blacksquare$  – from the analysis of the emission spectrum of targets) temperature of thermal spikes in pure iron (a) and aluminum (b), depending on the  $\text{Ar}^+$  ion energy. For the calculated curves  $T'=300$  K.

Calculated estimates of the temperature  $T^*$  in regions of thermalized cascades presented in Figure 3 and Table 1 were obtained, considering the following reasons.

In the thermalized dense cascade generated by an ion with energy  $E$ , the temperature increment (average energy per atom) is determined by:

$$E/N = 3/2 \cdot k \cdot \Delta T \quad (\Delta T = T^* - T'), \quad (2)$$

where  $N = \nu \cdot N_A$  is the number of atoms in a cascade ( $\nu = M/A$  is the number of moles of substance;  $M$  is the mass of the cascade,  $A$  is the atomic mass of a target); and  $N_A$  is the Avogadro's number.

Linear dimensions of a cascade  $\Delta R_{||}$  and  $\Delta R_{\perp}$  and its volume  $V(E) = 4/3 \cdot \pi \cdot \Delta R_{||} \cdot \Delta R_{\perp}^2$  were calculated using [8] (Table 1) and [8, 9] (Figure 1) algorithms.

As a result, the following relationship can be written for calculating thermal spike temperature  $T^*$ :

$$T^* = T' + \Delta T = T' + \frac{1}{2\pi} \cdot \left( \frac{E \cdot A}{k\rho \cdot \Delta R_{||} \cdot \Delta R_{\perp}^2 \cdot N_A} \right), \quad (3)$$

where  $T'$  is the initial temperature of the matrix (integrally heated target).

Estimates of the lower pressure limit in the regions of thermalized cascades (Table 1) were obtained on the basis of both experimental  $T$  and calculated  $T^*$  spike temperatures using the ratio [10, 11]:

$$P = \frac{E}{V} (c_p / c_v - 1). \quad (4)$$

Experimental  $V(E)$  and  $P$  estimates were obtained using the values of  $T$  (Table 1) and equations (2), (3), by substituting  $T^*$  for  $T$ .

Table 1 lists data on the theoretical [8] and experimental yield strengths for W, Fe, and Al. As the table suggests the pressure in the area of thermal spikes can reach values of several GPa, which is the confirmation of the probable emission of post-cascade solitary waves with the stresses at their fronts exceeding the yield strength of the material (including iron, aluminum and their alloys). Such waves may be responsible for the rearrangement of condensed matters under ion bombardment.

In [5, 6, 7], the experimental data on the rearrangement (including structural, phase, and interphase processes) of metastable media proceeding at an abnormally large depth much greater than the projected range of ions are given. As an explanation, hypotheses and models of processes self-propagating in metastable media are proposed, the rough analogy of which can serve the processes of

explosion crystallization of supercooled liquids and decomposition of supersaturated aqueous solutions during impact or shaking.

Figure 1 schematically shows the processes of diffusion-free fcc  $\rightarrow$  bcc phase transition in the Fe<sub>69</sub>Ni<sub>31</sub> alloy [6] and Al-4 wt. % Cu supersaturated solid solution decomposition [7] resulting from post-cascade shock waves effects in the volume of condensed matter. In [5], such processes are referred to the dynamic long-range effects or radiation-dynamic effects. The results of this study are proof of the principal possibility of considered effects.

The probable degree of the deviation of the state of thermalized regions from equilibrium one depends on various factors. It is determined by the energy of bombarding ions, the elemental composition of targets, thermalized region sizes arisen in them and the features of their evolution. It is necessary to consider separately the electronic and lattice processes [5].

The maximum contribution to the formation of emission spectra ( $\sim T^4$ ) is due to initial stages of the lifetime of thermal spikes.

The growth in the intensity of the IR wing of radiation correlates with an increase in the ion energy, ion current density, the irradiation fluence, and, as already noted, is associated with an integral heating of targets with ion beam (monitored via the thermocouple).

The data obtained for pure iron are in a good agreement with those of [3, 12]. The work [12] showed the presence of scattered atoms with thermal energies under irradiation of pure iron with 10-keV argon ions and using the time-of-flight techniques. The most probable velocity in the measured rate distribution, which appeared similar to the Maxwell distribution, corresponded to a thermal spike temperature of  $T = 5800$  K. It is shown that the majority atoms with thermal energies are neutral, since it does not feel any impact from the outside imposed electric field perpendicular to the surface with a field density up to 4 kV/cm.

Important evidence in favor of a special role of thermal spikes is the fact that the continuous emission (continuum) was not observed for the other methods of surface excitation.

Gas discharge, arc, spark, laser irradiation, and gas phase collisions of accelerated particles – all these factors generate only well resolved narrow lines corresponding to the chemical composition of targets [2], whereas only ion bombardment of solid bodies induces reliably observed continuum.

#### 4. Summary

It is shown that the continuous spectral bands in the range 400-700 nm, as part of the surface emission spectra of metal targets (Al, Fe, and W), in the course of their bombardment by Ar<sup>+</sup> 5-20 keV can be approximated by the Planck curves of thermal radiation. The temperatures derived from such approximation are in acceptable agreement with those simulated by Monte Carlo and Boltzmann-equation-based methods for the respective ion energies. Numerical models of the evolution of atomic displacement cascades to the stage of thermal spikes can explain the tendency to a decrease in the temperature of these spikes with increasing ion energy. In common it was shown the possibility for the direct experimental determination of the spike temperature (and, therefore, the density of energy release in cascade regions) by measuring the position of the maximum of the spectral density in the visible range. The pressures in the thermalized cascade regions in the studied metals are estimated for different ion energies.

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